

**WHAT IS CLAIMED IS:**

1. A non-linear optical active material for a non-linear optical device comprising:  
a matrix material;  
carbon nanotubes dispersed in the matrix material; and  
chromophores having non-linear optical properties attached to defect sites on the carbon nanotubes.
2. The material of claim 1, wherein the chromophores are selected from a group consisting of polymers, oligomers, monomers, dimers, organic molecules, atomic nanoclusters, nanowires, colloids and nanoparticles.
3. The material of claims 1 or 2, wherein the chromophores are chemisorbed to the defect sites on the carbon nanotubes.
4. The material of claims 1, 2 or 3 wherein the chromophores comprise organic dye molecules.
5. The material of claim 4, wherein the organic dye comprises a phenazine dye.
6. The material of claim 5, wherein the organic dye comprises PSF.
7. The material of claims 1, 2, 3, 4, 5 or 6, wherein the defect sites on the carbon nanotubes comprise a carboxyl group or a C<sub>1-6</sub> alkyl group.
8. The material of claim 7, wherein the C<sub>1-6</sub> alkyl group comprises a sec-butyl group.
9. The material of claims 1, 2, 3, 4, 5, 6, 7 or 8, wherein:  
the matrix material comprises a polymer matrix material; and  
the carbon nanotubes are well dispersed in the polymer matrix material.

10. The material of claim 9, wherein the polymer matrix material is selected from a group consisting of polyamide, polyester, polyurethane, polysulfonamide, polycarbonate, polyurea, polyphosphonoamide, polyarylate, polyimide, poly(amic ester), poly(ester amide), a poly(enaryloxynitrile) matrix or mixtures thereof.
11. The material of any one of claims 1 to 10 further comprising different types of chromophores attached to the carbon nanotubes, wherein the different types of chromophores have a peak sensitivity to different radiation wavelengths.
12. The material of any one of claims 1 to 11, wherein:  
the matrix material comprises a flexible thin film or a flexible fiber that is formed on a substrate; and  
an overall stiffness of the non-linear optical active material is determined by a stiffness of the substrate.
13. The material of any one of claims 1 to 12, wherein the carbon nanotubes are aligned in a controlled manner in the matrix material.
14. The material of claim 1, wherein the SuperNanoMolecular structures comprising the carbon nanotubes with attached chromophores are non-centrosymmetric.
15. The material of any one of claims 1 to 14, wherein the chromophores are covalently bound to a predetermined number of defect sites controllably arranged on the nanotubes.
16. The material of any one of claims 1 to 15, wherein the material has a controlled morphology.

17. A non-linear optical or electro-optical device comprising the non-linear optical active material of any one of claims 1 to 16.
18. The device of claim 17, wherein the device is selected from a group consisting of harmonic generators, frequency translation or mixing devices, optical memories, optical modulators, optical amplifiers, optical switches, directional couplers and waveguides.
19. The device of claim 17, wherein the non-linear optical active material is a thin film waveguide exhibiting a  $\chi^3$  effect.
20. The device of claim 17, wherein the non-linear optical active material is a thin film exhibiting a  $\chi^3$  effect incorporated into an optical switch.
21. The device of claim 17, wherein the non-linear optical active material is a thin film exhibiting a  $\chi^2$  effect incorporated into a device comprising thin film electrodes on the thin film, such that an optical beam can pass through the thin film and be deflected.
22. A method of making a non-linear optical active material, comprising:  
forming defect sites on carbon nanotubes;  
attaching chromophores having non-linear optical properties to the defect sites on the carbon nanotubes; and  
incorporating the nanotubes and the chromophores into a matrix material.
23. The method of claim 22, wherein the chromophores are chemisorbed to the defect sites on the carbon nanotubes.
24. The method of claims 22 or 23, wherein:

the step of forming defect sites comprises reacting the carbon nanotubes with an anionic initiator thereby generating anions on the surface of the carbon nanotubes; and

the step of attaching chromophores comprises covalently bonding the chromophores to the anions.

25. The method of claim 24, wherein the anionic initiator comprises an alkyllithium salt.

26. The method of claim 25, wherein the alkyllithium salt is *sec*-butyllithium which forms *sec*-butyl groups on the carbon nanotubes.

27. The method of claims 22 or 23, wherein:

the step of forming defect sites comprises reacting the carbon nanotubes with an acid thereby generating carboxyl groups on the surface of the carbon nanotubes; and

the step of attaching chromophores comprises covalently bonding the chromophores to the carboxyl groups.

28. The method of claim 27, wherein the acid comprises a mixture of sulfuric and nitric acids.

29. The method of any any one of claims 22 to 28, wherein the step of incorporating the nanotubes and the chromophores into a matrix material comprises incorporating the nanotubes and the chromophores into a polymer matrix material.

30. The method of claim 29, wherein the polymer matrix material comprises a flexible polymer thin film or a flexible polymer fiber.

31. The method of claim 30, wherein the step of incorporating the nanotubes and the chromophores into the polymer matrix material comprises incorporating the

carbon nanotubes and the attached chromophores into the polymer matrix material by interfacial polymerization.

32. The method of claim 31, wherein the polymer matrix material is selected from a group consisting of polyamide, polyester, polyurethane, polysulfonamide, polycarbonate, polyurea, polyphosphonoamide, polyarylate, polyimide, poly(amic ester), poly(ester amide), a poly(enaryloxynitrile) matrix or mixtures thereof.

33. The method of any one of claims 22 to 32, wherein the chromophores are selected from a group consisting of polymers, oligomers, monomers, dimers, organic molecules, atomic nanoclusters, nanowires, colloids and nanoparticles.

34. The method of claim 33, wherein the chromophores comprise organic dye molecules.

35. The method of claim 34, wherein the organic dye comprises PSF.

36. The method of claim 22, wherein the step of forming defect sites comprises controllably functionalizing the carbon nanotubes to controllably form the defect sites on the carbon nanotubes.

37. The method of claim 22, further comprising controlling a morphology of the non-linear optical active material.

38. The method of any one of claims 22 to 37, further comprising incorporating the non-linear optical active material into a non-linear optical device selected from a group consisting of harmonic generators, frequency translation or mixing devices, optical memories, optical modulators, optical amplifiers, optical switches, directional couplers and waveguides.